Tapered fibers embedded in silica aerogel

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We have embedded thin tapered fibers (with diameters down to 1 μm) in silica aerogel with low loss. The aerogel is rigid but behaves refractively like air, protecting the taper without disturbing light propagation along it. This enables a new class of fiber devices exploiting volume evanescent interactions with the aerogel itself or with dopants or gases in the pores. © 2009 Optical Society of America

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Silica aerogel is a nanostructured porous form of silica glass with extremely low density, typically more than 95% air but mechanically like a rigid solid [1]. The pores and connected silica network are structured on a scale of tens of nanometers, much smaller than the wavelength of light. The aerogel is therefore an effective medium of very low refractive index (typically between 1.01 and 1.05), albeit with some Rayleigh scattering. This "solid air" property makes it interesting in fiber optics, especially since fiber and aerogel are both made from silica glass and so are materially and thermally compatible. For example, embedding a fused coupler in aerogel should have little effect on its splitting characteristics, while providing a stable epoxy-free all-silica package suitable for harsh environments such as space. Thin single tapered fibers for nanowire circuits [2] or supercontinuum generation [3,4], where surface contamination is a serious problem, can be similarly protected. The volume evanescent interaction between tapered fiber and surrounding aerogel could be used for gas sensing because the aerogel is porous to gas and can be doped with fluorophores [5]. The aerogel can accommodate dopants that degrade at high temperatures, because the aerogel can be formed below 40°C. However, there are few reports of the use of aerogel in fiber optics. Tong *et al.* used it as a low-index substrate that avoids disturbing light propagation along tapered nanowires [2], but the fibers were simply laid on the aerogel, not embedded in it. They therefore remained exposed to air, (presumably) were not fixed in place, and so could become detached.

We report the formation of silica aerogel around the outside of tapered fibers, which become embedded in the aerogel. Sections of Corning SMF-28 fiber (cutoff wavelength of ~1260 nm) were tapered by heating and stretching in a small flame, and the taper waists were held suspended in air in open containers. The aerogel was made by one-step base-catalyst method [1]. Tetramethyl orthosilicate (TMOS), methanol, and water were mixed with an ammonia catalyst to form a fluid sol, which was poured into the container holding the tapered fiber. After a few minutes of gellation time, nanometer-scale colloidal particles of silica produced by the reaction connect together to form a rigid wet-gel or alcogel block (like the desired aerogel but with methanol in the pores) around the fiber.

The solvent was removed from the pores by supercritical drying, because capillary forces at interfaces between a liquid- and a vapor-phase solvent would collapse the porous gel structure during ordinary evaporative drying [1]. First the sample was immersed in liquid CO_2, which diffusively replaces the methanol in the gel. We did this because CO_2 has a lower critical temperature T_c (31.1°C) than methanol (240°C) and is not flammable. Then the sample was pressurized above the critical pressure, heated above T_c, then gradually depressurized; see Fig. 1. The result was an aerogel block with an embedded tapered fiber; see Fig. 2.

Shrinkage of the gel as it forms causes stress that can break the thin tapered fiber or crack the gel body. For example, the sample in Fig. 2(a) cracked and shrunk by ~15% (in linear dimension), and the 10 μm diameter tapered fiber inside was broken. To minimize shrinkage and prevent breaks, we optimized parameters such as how the fiber is held, the ratio of the chemicals, the gelling time, and the supercritical drying cycle [6]. The fibers were usually held taut, but very thin ones (1 μm diameter) were slackened before immersion to reduce the stress to which they could be subjected; see Fig. 3(c). Our most

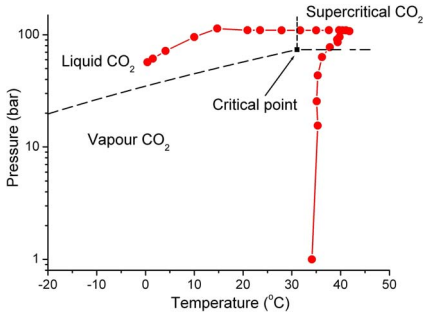

Fig. 1. (Color online) Measured supercritical drying cycle on a CO_2 phase diagram. The pressure and temperature were recorded every 5 min and marked by dots. The liquid-vapor phase boundary is not crossed; the liquid and vapor phases are never present at the same time.

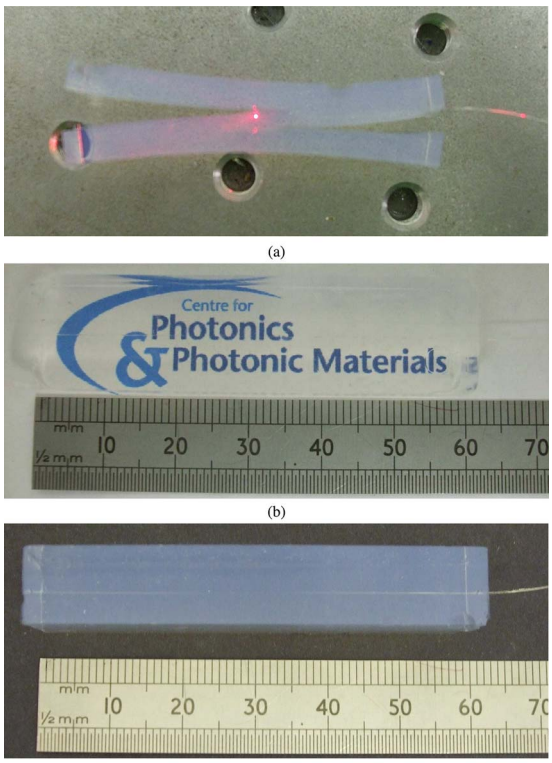

Fig. 2. (Color online) Photos, to the same scale, of (a) a cracked aerogel sample with a broken embedded taper carrying some red laser light, (b) front and (c) side views of a good sample against different backgrounds.

successful sol was made by mixing TMOS, methanol, water, and ammonia with a molar ratio of 1:3:4:0.0028. We quickly placed covers on the fiber containers after filling with sol to prevent ambient drying. The sol turned to wet gel in a few minutes, was left to age for two days, then was soaked in methanol for two days to remove excess water by diffusion. The samples were immersed for 6 h in liquid CO_2 in an autoclave at approximately 55 bars pressure to exchange the methanol for liquid CO_2; they were then supercritically dried along the path shown in Fig. 1.

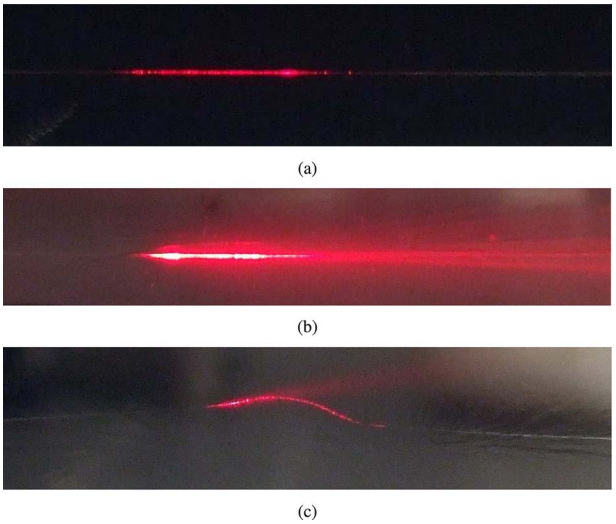

Fig. 3. (Color online) Photos of embedded tapered fibers with waist diameters of (a) 10 μm, (b) 2 μm, and (c) 1 μm. The fibers carry red laser light from left to right. Noticeable scattering begins at the start of the 1-cm-long waist. The paths of the scattered light through the aerogel can be seen. The images are ~3.1 cm long.

Aerogel made this way is brittle but quite robust and could be handled normally by hand without damaging the aerogel or the embedded fiber. However, it is hygroscopic, because its large internal surface area is covered with hydroxyl groups [7], decomposing immediately on contact with liquid water. It also gradually shrinks as it absorbs water vapor from the air, which can eventually break an embedded fiber. To prevent this, we stored the samples in desiccant boxes. With these precautions the aerogel shrunk in length by only ~4% during drying and could remain intact with the fiber inside unbroken. The aerogel was barely visible against a light background [Fig. 2(b)] and had a faint blue color from Rayleigh scattering against a dark background [Fig. 2(c)]. A refractive index of $n=1.048\pm0.005$ at 633 nm wavelength was determined by measuring the critical angle within the sample. The index variation with wavelength, related to the suitably scaled dispersion of bulk silica, is very small [8] and irrelevant to our results. Its measured density of $\rho=0.24$ g/cm^3 was consistent with the approximate relation $n=1+0.21\ \rho$ (g/cm^3) [9]. The aerogel was permeable to gas; the weight of centimeter-scale samples changed in heavy gas such as SF_6 and butane, with an ~10 s time constant consistent with other reports of gas diffusion in aerogel monoliths [5] and only ~10× slower than unimpeded diffusion in air.

The losses of tapered fibers with waists of 10 mm length and diameters of 10, 2, and 1 μm were measured by the cutback method with a large-area photodiode and a 1310 nm laser. This gives the loss of the entire tapered fiber, including the 10-mm-long waist and the taper transitions. The initial losses were all <0.1 dB immediately after the tapers were made, which is typical of tapered fibers in air [10]. After embedding in aerogel the losses became 0.12, 0.31, and 1.14 dB, respectively, for the three waist diameters. The fraction of mode power in the evanescent field around a tapered fiber [10] also increases with decreasing diameter (we calculate it to be 0.0003, 0.03, and 0.2, respectively, for our tapers [11]). Much of the loss is therefore probably scattering of this evanescent field by the aerogel, which Fig. 2(c) shows is a scattering medium. However, the surrounding aerogel prevented further scattering loss over time due to the settling of dust. This process quickly degrades unprotected thin tapered fibers such as like those that have been used for supercontinuum generation [3,4] or so-called nanowires or microfibers [2].

The trend in scattering loss was visualized with red laser light, which scattered more at the 2 μm waist [Fig. 3(b)], than at the 10 μm waist [Fig. 3(a)]. However, the visual effect was exaggerated by higher-order modes with stronger evanescent fields (the fiber was multimode for red light). Even more light was scattered from the 1 μm waist [Fig. 3(c)],

where we decreased the laser power to avoid saturating the image.

Loss spectra measured with an optical spectrum analyzer (OSA) and broadband LED source for 10 and 1 μm waists in air and in aerogel are shown in Fig. 4. All show large OH peaks at ~1383 nm owing to diffusion, into the glass, of water produced by the combustion of butane in the taper rig's flame [12]. The additional loss due to the aerogel is consistent with the single-wavelength data (which are more accurate because light capture by the OSA is less reproducible) and is approximately spectrally flat away from the OH peaks. We believe that an increased evanescent field at longer wavelengths is counteracted by reduced Rayleigh scattering. However, for the 1 μm waist in aerogel there is a second narrower loss peak at 1366 nm, corresponding to the first overtone of free SiOH groups at the surfaces inside the aerogel [7]. When the aerogel was exposed to ambient air for 3 h, this peak decreased and the weight of the aerogel increased by 3.2%. Our interpretation is that the hygroscopic aerogel absorbed moisture from the air, converting the free SiOH to hydrogen-bonded SiOH, as observed by Miner *et al.* [7].

This sensitivity to water highlights potential applications for gas sensing; the nanoporous aerogel protects the taper from particulate contamination while allowing gas to diffuse to the evanescent field around the taper. However, hygroscopic materials are obviously not stable in most environments, so in applications it is important for the aerogel to be hydrophobic. We have produced hydrophobic aerogels by chemically replacing the surface hydroxyl groups with trimethylsilyl groups before drying [13]. As well as making the aerogels waterproof (they can be immersed in water for hours without effect), the process also reduced shrinkage in length to <1%. Indeed, hydrophobic aerogel has been reported stable for a year while floating on water [14]. Embedding of submicrometer tapered fibers in hydrophobic aerogel is under investigation.

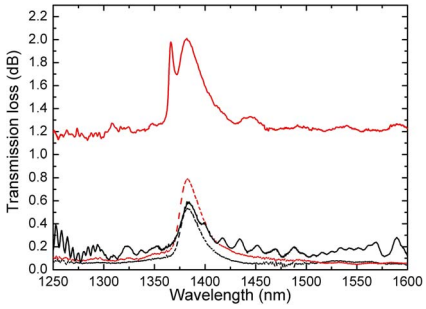

Fig. 4. (Color online) Transmission spectra of tapered fibers in air (broken curve) and in aerogel (solid curve) for waist diameters of 10 μm (darker) and 1 μm (lighter).

In conclusion, we successfully embedded narrow tapered fibers in silica aerogel. The measured losses were ~0.1, 0.3, and 1.1 dB for waist diameters of 10, 2, and 1 μm, respectively. Low-loss encapsulation in aerogel protects thin tapered fibers from contamination while allowing gases to pass and should enable a new class of fiber devices exploiting volume evanescent interactions with the aerogel or with dopants or gases inside it.

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